AIR-COUPLED ULTRASONIC LAMB WAVES FOR PROCESS MONITORING

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Abstract

The production of foam-metal composites is always a “hidden” process as the metal skin denies access to the chemical reactions of the core. This reaction (formation of a foam, polymerization, bonding to the skins) is crucial for the quality of the product. Current measurement techniques are not capable of monitoring these processes in a realistic setting. We present a new approach to probe the behaviour of the reactive mixture and its transition to a fluid and later solid foam, by using an air-coupled ultrasonic plate wave (“Lamb wave”) in the substrate. The methodology is introduced along with a case study of its application for monitoring of a similar process of coating drying (fluids, paint) on a steel substrate.

Introduction

Sandwich structures consisting of a foam core and metal or composite skins find widespread use because they combine structural strength with good thermal insulation at a light weight. Aerospace carbon components are usually produced using pre-made blocks of foam as core with the epoxy matrix functioning as glue between core and skins. The key requirement for steel-foam sandwich elements (SFSE) is a cost-effective production process which leads to a polyurethane (PUR) core foamed in-situ between two steel sheet skins. The PUR supplies the core as well as the glue for bonding to the skins. The chemical and physical processes (foaming and polymerization) at the steel surface are critical for the quality of the final element - poor adhesion will lead to delaminations under thermal load (day-night-cycle) and costly replacement of the panel. While there are established methods to study the dynamics of a free foam (“Fomat”, [1]) and the chemical reactions (e.g. FTIR-spectroscopy, [2]), currently none is capable of monitoring the processes at the boundary to the “natural” substrate of the foam, the steel sheet. A similar process also happening on the surface of a steel substrate is the drying of paint or any other fluid coating.

Both types of boundary-layer processes can be probed by using a low-frequency plate acoustic wave (PAW), thus making the steel substrate itself the sensor for monitoring the physico-chemical phenomena in its proximity. To avoid any influence by the introduction of activation heat (or its removal by a heat sink), we use a non-contact excitation/detection of the PAW by air-coupled ultrasound (ACU).
Methodology

The selective excitation of plate waves by air-coupled ultrasound (ACU) in the slanted mode is the basis of all presented experiments. The incident angle of the ACU beam can be adjusted until its pressure distribution on the plate surface matches the wavelength of the lowest order antisymmetric \( a_o \) plate wave (see Fig. 1). The conversion of ACU into the guided wave is optimal at the so-called resonance angle \( \Theta_0 \) defined by:

\[
\sin \Theta_0 = \frac{v_{ACU}}{v_{PAW}},
\]

where \( v_{ACU} \) and \( v_{PAW} \) are the respective phase velocities. This resonance mode conversion can be used in the focused slanted transmission mode (FSTM) or in the single-sided focused slanted reflection mode (FSRM, see Fig. 1): After a propagation distance of a few centimetres the plate wave is detected by a second transducer; the waveform is digitized and stored. A discrete Fourier transform (DFT) recovers the amplitude and phase of the output signal, even if it is very week.

A similar setup has previously been used for non-destructive testing and imaging [3] as well as precise measurement of plate and surface wave velocities [4]. In the thin-plate limit, the phase velocity of the \( a_o \)-mode of a plate wave can be approximated as [5]:

\[
v_{a_o} = \sqrt{\frac{E}{3\rho(1-\mu^2)}} \sqrt{\frac{\omega D}{2}}.
\]

It is determined by the Young’s modulus \( E \), mass density \( \rho \), Poisson’s ratio \( \mu \), the circular frequency \( \omega = 2\pi f \) and the plate thickness \( D \). Any perturbations in the mechanical state of the substrate (e.g. thin coatings) can be considered by contributions to the stiffness, mass density or thickness thereby changing them to effective values. With \( m \) the mass per unit area of the undisturbed substrate plate and \( \Delta m \) the additional mass of the coating, the plate wave velocity changes as [6]:

\[
v_{a_o} \sim \sqrt{\frac{1}{m + \Delta m}}.
\]

So a thin fluid layer will contribute to the mass, while no change to the bending modulus of the plate is expected, thus reducing the plate wave velocity. Changes in the velocity \( v \) are detected by tracking the phase of the received ultrasonic signal. By introducing the velocity variation \( \Delta v = v' - v \) and the corresponding wave numbers \( k = \omega / v \) and \( k' = \omega / v' \), for the phase shift \( \Delta \phi \) along the path length \( x \) we obtain:

\[
\Delta \phi = (k' - k)x = \left( \frac{\omega}{v + \Delta v} - \frac{\omega}{v} \right)x = \left( -\frac{\omega \Delta v}{v^2 + v \Delta v} \right)x.
\]
Assuming a small velocity change ($\Delta v \ll v$) the velocity change can be calculated from the measured phase shift:

$$\Delta v = -\frac{v^2}{\alpha x} \Delta \varphi .$$

(5)

The experiments were carried out on steel sheets ($D = 0.95 \text{ mm}, v s \approx 1200 \text{ m/s}$) using 200 kHz ACU and a propagation path length $x = 10 \text{ cm}$. In a normal lab environment, the phase of the received signal recovered by DFT is accurate by approximately 10°, which means the threshold for a detectible velocity variation is $\Delta v / v \approx 0.2\%$. Sampling can be acquired at rates up to 20 per second.

### Experiments and Results

#### Solvents and Paint on a Steel Substrate

To study the influence of thin liquid coatings on the PAW propagation, the steel sheets were coated by three substances: Tap water (some detergent added to aid the surface wetting), the organic solvent isopropyl alcohol and a water-based paint (model paint designed to approximate the type used in the automotive industry, supplied by F. Brinckmann, TU Darmstadt). A phase-tracking measurement always starts before applying the coating to secure a clear starting point for the undisturbed substrate. In the case of the solvents a fluid layer of ~0.4 mm was created by pouring 1.5 ml of liquid onto a walled-off area of the substrate. Figure 2 shows the phase curves for drying of water and isopropyl. At the time of wetting the phase drops by approximately 40° and then recovers almost linearly over time until reaching the initial value. As expected the process takes much longer (~3h) for water than for the more volatile isopropyl (~45min). The departure from a linear recovery after some time is easily explained: at this point the film no longer covers the full area, and the active surface for evaporation reduces.

![Figure 2: FSRM phase curves for water (left) and isopropyl (right) wetting/drying on steel substrates. Measured phase values shown as dots, moving-window-averaging as solid lines and linear fit of the early recovery rate.](image)

To obtain a thin layer (~0.1 mm) of the viscous automotive paint, it has to be mechanically spread onto the surface using a special device (Erichsen “Model 360 fixed gap film applicator”). To avoid disturbing the ACU setup, the steel substrate was thoroughly fixed in a specially made frame holder (see Fig. 3).
Figure 3: Setup for ACU phase measurements. Left: Specimen holder with visible ultrasound transducers. Centre: Steel substrate inserted. Right: After application of fresh paint.

The resulting phase curve (Fig. 4) features a sharp drop at the start and a slow recovery similar to the experiments described above for the simpler solvents. However, at least two regimes of different recovery rate (see straight lines included in the figure) are seen in this case. The steeper phase rise within the first two hours is caused by the additional butyl glycol content of the paint (evaporating faster than water); the subsequent slower recovery \((t > 5h)\) is due to delayed diffusion of the remaining water within the \(~100 \, \mu m\) thick layer. The process is still not finished after the time period of 7.5 h shown here and the paint remains soft and sticky. In an automotive application, the painted part would be pre-dried at 70°C (hot air) before sealing with a clear protective coating and final curing at 140°C.

Figure 4: Measured phase variation during application and drying of water-based automotive paint.

**Polyurethane Foam on a Steel Substrate**

Polyurethane (PUR) foams are cellular, expanded materials synthesized by the polyaddition reaction of diisocynate and polyol in the presence of additives such as blowing agents, catalysts, chain extenders, branching agents or fillers and pigments. Figure 5 (left) gives a schematic representation of the exothermic polyaddition reaction of diisocyanate and polyol to polyurethane. Because most (about 80 % [7]) of the polyurethanes are used as foams blowing agents are indispensable. They can either be chemical, e. g. water or physical blowing agents, e. g. carbon dioxide or low boiling solvents like halogen derivates as e. g. chlorofluorocarbons (CFCs). Water reacts with isocyanate groups to produce carbon dioxide which serves as foaming agent (Fig. E, right). As the use of physical blowing agents requires sophisticated process technology which cannot be supplied in a lab environment, only water-driven foams were used.
Steel foam sandwich element (SFSE) structures can either be made in a continuous or a discontinuous process. Most sandwich structures are produced in a continuous way in a double-belt press. The reaction mixture is inserted into the space between two constantly moving rigid or flexible skins. Figure 6 gives a schematic representation of an industrial production line.

To simulate an industrial-style production of SFSE in the lab, a compact (140x140x100 mm) foaming cell for manufacturing test specimens (adhesive strength, based on DIN 53292) was constructed. It includes a massive aluminium foaming chamber with paper inlays (for easy foam removal) and the steel substrate forming the bottom (see Fig. 7).
A small window provides access of the ultrasound apparatus to the steel sheet. It is framed by magnets to clamp down the metal, thus reducing possible motion. The requirements of destructive testing defined the dimensions of the substrate with a plate wave propagation path of about 50 mm. As the chemical reaction of the foam generates enough pressure to deform slightly the 0.75 mm steel plate, an analogue-optical distance correction loop was installed to keep the propagation paths transmitter-plate / plate-receiver in air constant. Acquisition of the ultrasonic signal always started on the undisturbed substrate before adding the reactive mix in a liquid state, just before the foaming process starts. Development of a solid PUR foam is always governed by the dynamics of two parallel reactions: release of CO₂ (foaming) and the formation of a cross-linked polymer network. In an open-beaker foaming process the volume increase can be best tracked by following the level of the material within the container (See Fig. 8). After a short dormant phase the volume increases rapidly. To ensure that the material can fill all voids the chemistry is usually set in a way that the polymer matrix is still fluid during the volume increase. The process is concluded by the polymerization, leading to a light, solid foam.

As a result, three major phases of the process are expected: Dense fluid reactive mix (ρ~1 g/cm³), light liquid foam (ρ=30-40 kg/m³) and a stiff finished foam of the same low density. While the first (fluid) and the third (solid) phase are indicated clearly in the air-coupled Lamb wave measurement, the second phase (liquid foam) can be seen only as a transition between the other two or is indistinguishable from the solid final result (see Figs. 9, 10). The amplitude of the received ACU shows variation only during the first few seconds after the PUR is added to the chamber. As soon as the liquid mix touches the substrate, the amplitude drops sharply (see Fig. 9). As the reaction starts and a light liquid foam develops, the amplitude recovers quickly and stays at a steady level. Such an amplitude behaviour can be attributed to the energy exchange in the layered structure: An efficient energy “leakage” in the for of a bulk wave radiated into a liquid (“fast” medium-density medium, good acoustic contact) changes for low attenuation caused by the interface-dominated viscoelastic interaction with a low-density, low-velocity polymer foam. There is no distinct difference between liquid and solid foam in the ACU amplitude.
Figure 10 shows the phase variation of the PAW during foaming. When the liquid reactive mixture is first deposited onto the plate, the phase exhibits a negative shift (lower wave velocity due to extra mass). As the PUR forms a liquid foam of a low density and stiffness, the phase recovers and then, as the foam hardens, reaches a plateau above the initial value (higher velocity due to stiffening). The “ripples” in the phase curves are mainly due to the finite time constant of the distance feedback loop, always allowing for a small displacement of the steel sheet before taking a correction step.

The goal of all process monitoring efforts is a prediction of the final product quality. The critical property is the adhesive strength of the interface (normal tension necessary for element destruction). The adhesive strength depends on the surface preparation of the substrate (coatings, activation, contaminations), the polyurethane mixture and the process parameters. A change in these parameters is expected to affect the ACU amplitude and phase curves. Subsequent comparison with the results of destructive tension testing will provide a correlation between the development of the different phases of the foaming reaction (measured remotely by ACU) and the eventual adhesive strength. These measurements with systematic variation of the PUR process parameters are being currently conducted at the LFK.

Preliminary results show an adhesive strength of 2 - 20 N/cm$^2$ (20 - 200 kPa) of the steel-foam interface with a purely cohesive fracture (within the foam). The fracture typically follows the steel surface in the first or second foam cell layer for some distance before then sometimes turning into the core volume (see Fig. 11). As seen in Fig I, the stress-strain relation of the foam is almost linear with some stiffening with increasing strain. The failure itself is brittle without visible load redistribution (see force-displacement diagram, Fig. 12).

Fig. 11: Photo of a finished SFSE showing the steel substrate (left), the inside of the central plate (middle) and the corresponding core (right) after tensile testing.
Summary

ACU with plate wave mode conversion has been shown to provide a remote, non-contact non-destructive means for monitoring reaction processes on plate-like substrates. The amplitude and phase measurements reveal non-invasively the dynamics of drying coatings (solvents, paint). The ACU plate wave sensor has been integrated in a compact laboratory foaming cell for a production of SFSE that simulates the large-scale industrial production process of insulation and construction panels. The methodology has been shown to be capable of tracking the foaming process at the metal-foam interface from the exterior of the SFSE.

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References